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A conjecture on the Process of Charge Carrier Separation in Columnar Structure of Material and at Interfaces

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1. Introduction

The problem of charge carrier separation in insulators or semiconductors is an important topic which has many fundamental and practical implications. In semiconductors, it relates to the problem of conversion efficiency of photocells, to the injection process at the metal-semiconductor interface and to the application of semiconductors for the detection of high energy radiation; in insulators, the understanding of this problem is important for their application in high electric fields, for their use as detection media in radiation detectors, and for an understanding of the separation of a charge carrier pair in the condensed phase.

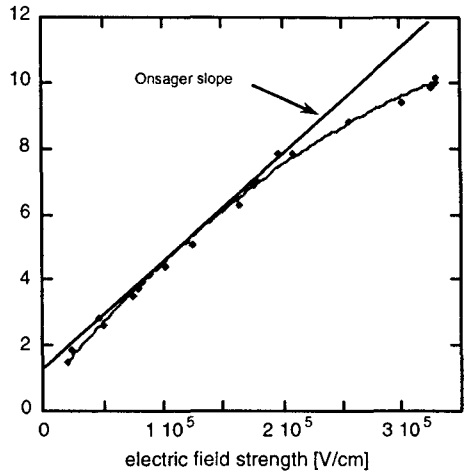
2. Charge carrier separation in polymers and insulating liquids

Theoretically, the problem of the migration of a charge carrier pair of opposite polarity in their mutual attractive Coulomb field and under the influence of temperature and an externally applied field has been solved by Onsager [Onsager, 1938]. A critical separation distance, r_c , is defined at which the Coulomb energy of attraction is equal to the thermal energy k_BT,

$$r_c = \frac{e_0^2}{4\pi\varepsilon_0\varepsilon_r k_B T}$$

Here, e_0 denotes the electron charge, ε_r is the relative dielectric constant, ε_0 is the permittiviy of the vacuum. If the initial separation of the charge carrier pair is much greater than r_c then diffusion is prevailing and the pair escapes ist mutual attraction. If the initial separation is much smaller than r_c then the prevailing Coulomb attractions leads to recombination. A presumption of the Onsager model is the requirement that the characteristic length of the charge carrier drift (jump length or scattering length) is small compared to r_c. This condition is usually fulfilled for some liquid hydrocarbons] or polymers since the mobility of the charge carriers involved is small (i.e. $<< 1 cm^2V^{-1}s^{-1}$).

Charge carrier pairs in insulators can be produced by irradiation of the material with γ-rays or fast electrons. An experimental test of the applicability of the Onsager model consists in the measurement of the radiation-induced current as a function of the applied electric field strength An example of the ionization current in poly-vinylcarbazole is shown if Fig. 1.



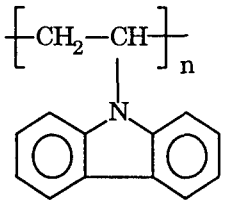


Fig. 1: Electron/positive ion yield for x-irradiation of poly-vinycarbazole , PVC, (after [Hughes, 1971a])

The initial rise of the current with field is followed by a range where the current is a linear function of the applied electric field strength. From the Onsager model a unequivocal value for the slope (sl) of this line divided by the intercept (in) at E=0 follows which depends on the macroscopic properties of the medium only,

$$sl / in = \frac{e_0^3}{8\pi\varepsilon_0\varepsilon_r k_B^2 T^2}$$

Besides for PVC, measured currents in anthracen crystals [Hughes, 1971b] and in polyethylene terephthalate [Maeda, 1979] were in satisfactory agreement with the theoretical prediction for sl/in. In many other polymers (as for instance polyethylene), the radiation induced ionization currents do not follow the Onsager model, probably due to the fact that permanent trapping of the charge carriers in the bulk of the material occurs. Proof for this assumption are the thermally stimulated currents observed in these polymers after irradiation (e.g. [Lapke, 1982]) and the complicated dependence of the induced current on the dose rate at a given field strength [Schmidt, 1994]. In order to observe an Onsager-type behavior, at least one type of charge carrier has to exhibit a mobility which allows it to reach the collecting electrode or its counter ion. In many insulating liquids where both types of charge carriers are mobile, the Onsager model for charge carrier escape desribed the measured ionization currents correctly [Schmidt, 1997].

3. Charge carrier separation in discotic liquid crystals

Discotic liquid crystals are characterized by a spatial order of the molecules comprising the crystal. A simpliefied representation is shown in Fig. 2.

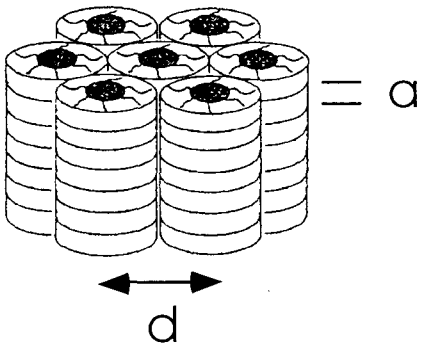


Fig. 2: Stacks of liquid crystal molecules in the discotic phase (a – distance between adjacent molecules in the stack; d – distance between the columns)

Here, the positive charge carrier (hole) preferentially moves along the direction of the columns, inter-columnar transfer is not frequent since the distance a is much smaller than the separation of the columns, d.

It may be speculated that the one-dimensional movement of the hole would have an effect on the charge carrier separation process. In our previous work [Nakayama, 1999] we found that the escape of charge carrier pairs generated by photoionization in discotic liquid followed the Onsager model. The charge collected as a function of applied field strength is shown in Fig. 3. The measured sl/in – value agreed very well with the theoretical expectation. From this result, we have to assume that inter-columnar transfer of the positive charge and the negative charge (and if we assume that the negative charge migrates along the stack, too) can take place. Since the quantum yield for charge carrier production is low, we have to assume that the mean initial separation distance is small, of the order of 2 or 3 nm. The Coulomb-field at such a sepaaration distance is greater than 10^6 V/cm. The influence of the electric field on the transport properties of the hopping charge carriers has to be taken into account. It was found that high electric fields lead to
an increase of the hopping mobility given as [Bagley, 1970],
In other words, the barrier hindering the inter-columnar transfer of charges is reduced by the high Coulomb field of attraction.

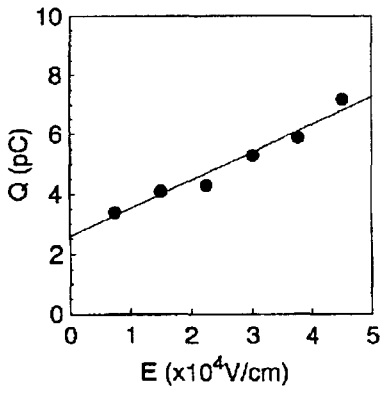


Fig. 3: Field dependence of the collected positive charge due to hole migration in the discotic phase of HHOTP [Nakayama, 1999]

4. Positive charge injection into discotic liquid crystals at the interface

The influence of one-dimensionality on the charge carrier separation process could be observed if holes could be injected from an electrode. The separation would be between the injected hole and its image charge in the electrode. This problem has been studied in detail by the process of electron injection into insulating liquids by photoeffect [Schmidt, 1997]. In this case, a proportional relationship between photo current (or collected charge) and the applied electric field would be expected. Such experiments need to be done.

On the other hand, measurements of the transverse photo conductivity of discotic liquid crystals could give insight into the inter-columnar charge transfer process.

5. Conclusion

The experimental investigations of the process of charge carrier separation and of the transport properties of the charge carriers enhance our understanding of the processes influencing the quantum yield of photo-electric devices based on liquid crystals.

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